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Triple microporous layer coated gas diffusion layer for performance enhancement of polymer electrolyte fuel cells under both low and high humidity conditions

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HIGHLIGHTS

- Novel triple MPL coated GDL was developed to enhance the PEFC performance.
- The thin hydrophilic layer coated on the hydrophobic double MPL is effective at conserving MEA hydration under low humidity.
- The triple MPL with an appropriate hydrophobicity gradient was effective at reducing flooding under high humidity.

ARTICLE INFO

ABSTRACT

Enhancement of the performance of polymer electrolyte fuel cells (PEFCs) requires an appropriate water balance between the conservation of membrane humidity and the discharge of excess water produced in the cell. In the present study, a novel triple microporous layer (MPL) coated gas diffusion layer (GDL), in which a hydrophilic layer was coated on a hydrophobic double MPL, was developed to enhance the PEFC performance under both low and high humidity. The thin hydrophilic layer in the triple MPL is effective at conserving the humidity of the membrane electrode assembly (MEA) under low humidity, while the hydrophobic double MPL between the hydrophilic layer and the carbon paper substrate prevents removal of water from the hydrophilic layer. This results in a significant enhancement of the ability of the GDL to prevent dehydration of the MEA. The triple MPL coated GDL, where the polytetrafluoroethylene (PTFE) content in the hydrophobic MPL in contact with the hydrophilic layer is set to 30 mass% and that in contact with the substrate is set to 10 mass%, is effective at expelling excess water from the catalyst layer, which results in much higher PEFC performance under high humidity than that for a conventional hydrophobic MPL coated GDL.

1. Introduction

Polymer electrolyte fuel cells (PEFCs) generally have external humidifiers to supply humidified fuel and oxidant gases, which prevents dehydration of the membrane electrode assembly (MEA). However, such additional humidifiers make the PEFC system complex and increase the production cost. The humidification requirements of the anode and cathode are different. At the anode, it is possible to introduce humidified hydrogen gas using water generated during the reforming process in fuel production. Furthermore, humidified pure hydrogen gas that is not used for the electrochemical reaction can be recirculated in the PEFC system. It is therefore possible to remove external humidification at the anode. At the cathode, air, from which oxygen is consumed in the electrochemical reaction, is usually exhausted without recirculation. Therefore, the humidified air is generally supplied using external humidification. If a PEFC could be operated without cathode humidification, then external humidifiers could be removed, which would result in a very simplified PEFC system with increased total efficiency and reduced cost. Therefore, one of the most important issues to advance the commercial viability of PEFCs is the development of high performance PEFCs that can operate without cathode humidification [1–5].

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The design parameters for the gas diffusion layer (GDL) in the PEFC, such as its pore size, thickness and hydrophobic and hydrophilic properties, influence the water management characteristics during PEFC operation. Several investigations have demonstrated that a hydrophobic microporous layer (MPL) coated on a GDL substrate can effectively improve the water management characteristics and thereby enhance the PEFC performance [6–13]. An appropriate MPL coated GDL prevents dehydration of the MEA under low humidity conditions and reduces flooding under high humidity conditions. However, the appropriate design parameters for the MPL coated GDL are different under low and high humidity conditions [14]. An MPL coated GDL designed to prevent dehydration of the MEA under low humidity conditions is generally inferior at reducing flooding under high humidity conditions. Even when a PEFC is operated under low humidity conditions, it is essential to prevent flooding due to water produced in the cell at high current densities. Therefore, it is important to develop a robust MPL coated GDL that can enhance the PEFC performance under both low and high humidity conditions.

Although the hydrophobic MPL coated GDLs have been commonly used to improve the water management properties of PEFCs, some studies have reported that either the hydrophilic MPL [15,16] or hydrophilic treatment of the GDL substrate [17] are effective to improve the water management properties of PEFCs, thereby enhancing the PEFC performance. The authors have also reported that the hydrophilic MPL coated GDL using polyvinyl alcohol (PVA) was effective to enhance the PEFC performance under low humidity conditions [18]. Although the ability of the hydrophilic MPL coated GDL to conserve the humidity of the MEA was enhanced, it was relatively easy for water in the hydrophilic MPL to be expelled to dry air in the substrate. Therefore, enhancement in the PEFC performance was not significant when using the hydrophilic MPL coated GDL. Thus the authors have developed a new hydrophilic and hydrophobic double MPL coated GDL [18]. A thin hydrophilic layer using either PVA or titanium dioxide (TiO$_2$) coated on the hydrophobic MPL was effective to conserve the humidity of the MEA under low humidity conditions, while the hydrophobic intermediate MPL between the hydrophilic layer and the carbon paper substrate prevented the removal of water from the hydrophilic layer. This resulted in a significant enhancement of the PEFC performance under low humidity conditions, compared with that for a hydrophilic MPL coated GDL. Under high humidity conditions, the double MPL coated GDL with the hydrophilic layer using PVA increased the water accumulation at the cathode catalyst layer, which increased the concentration overpotential and thereby lowered the PEFC performance. However, the double MPL coated GDL with the hydrophilic layer using TiO$_2$ was effective to reduce flooding under high humidity conditions, which resulted in higher PEFC performance than that for the conventional hydrophobic MPL coated GDL [19].

In the present study, a novel triple MPL coated GDL was developed, in which the hydrophilic layer was coated on a hydrophobic double MPL. The hydrophobic double MPL in the triple MPL coated GDL had a gradient of hydrophobicity due to variation of the polytetrafluoroethylene (PTFE) content, where the PTFE content in the hydrophobic MPL in contact with the hydrophilic layer was set to 30 mass% and that in contact with the substrate was set to either 10 or 40 mass%. The triple MPL was evaluated to determine the optimal parameters for promoting the discharge of excess water from the catalyst layer and thereby enhancing the PEFC performance under both low and high humidity conditions.

2. Experimental

2.1. Tested GDLs

The GDL used at the anode was a commercial carbon paper without an MPL (SGL SIGRACET® 24BA), which was loaded with 5 mass% PTFE to impart hydrophobicity [8]. The SGL24BA GDL has a thickness of 190 μm, an areal weight of 54 g m^{-2}, and 84% porosity. Fig. 1 shows hydrophobic MPL, double MPL and triple MPL coated GDLs used at the cathode. The hydrophobic MPL coated GDL consisted of a carbon paper substrate (SGL24BA) coated with an MPL of 30 mass% PTFE and carbon black. The hydrophobic MPL was coated on the substrate as follows. A slurry containing a PTFE dispersion liquid (Daikin Polyflon D-210C), carbon black (Denki Kagaku Kogyo DENKA BLACK® HS-100), distilled water and a surface-active agent was mixed using an impeller blade-type mixer, and then spread on the substrate using a bar coating machine. The MPL was dried in an oven and then heated at 350 °C to remove any remaining solvent and surfactant, sintering PTFE and carbon black onto the substrate. For both the double MPL and the triple MPL coated GDLs, a hydrophilic layer of 25 mass% TiO$_2$, 5 mass% silicone, and carbon black was coated on the hydrophobic MPL coated GDL [19]. The PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDL was varied between 10 and 40 mass%. For the triple MPL coated GDL, the PTFE content in the hydrophobic MPL in contact with the hydrophilic layer was set to 30 mass% and that in contact with the carbon paper substrate was set to either 10 or 40 mass%.

2.2. PEFC performance tests

A commercial MEA (GORE PRIMEA®5580, membrane thickness: 30 μm, Pt loadings at the anode and cathode: 0.4 mg cm^{-2}) was used in the PEFC performance tests under low and high humidity conditions. The active area of the MEA was 4.2 cm^2. A separator with a triple serpentine flow channel configuration was used, and the anode and cathode gases were supplied in a counter-flow orientation. The cell temperature was set at 75 °C. Hydrogen and air utilization was set to 70 and 60%, respectively. For PEFC performance tests under low humidity conditions, the relative humidity (RH) of the gas supplied to the cathode was set to 0%, while that supplied to the anode was maintained at 60% RH. For PEFC performance tests under high humidity conditions, the RH of the gases supplied to both the anode and cathode was set to 100%. The
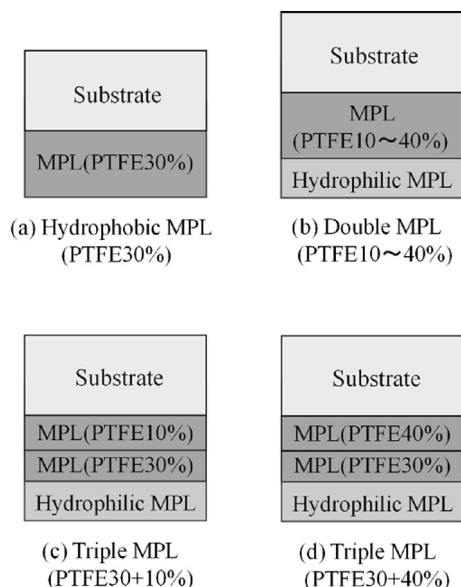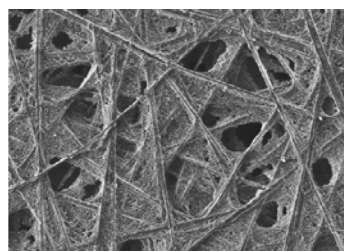

Fig. 1. Hydrophobic MPL, double MPL and triple MPL coated GDLs used at the cathode.

IR (ohmic loss), activation and concentration overpotentials were measured separately [14]. There was no significant difference in the activation overpotentials obtained for all the MPL coated GDLs. Therefore, the influence of the MPL coated GDLs on the IR and concentration overpotentials are discussed with the following test results.
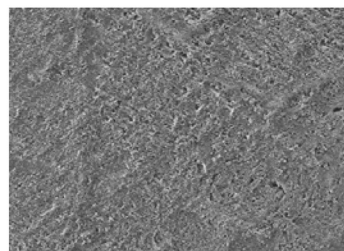
3. Results and discussion

3.1. Pore diameter, thickness, contact angle and air permeability of the tested GDLs

Fig. 2 shows surface scanning electron microscopy (SEM) micrographs of the GDL substrate (SGL24BA), the hydrophobic MPL and hydrophilic MPL in the triple MPL coated GDL. The substrate had a mean flow pore diameter d_m of 42 μm. The mean flow pore diameter of the GDL was measured by the through-plane permeability test using the wetted GDL, in which a low surface tension liquid [20] with a contact angle of 0° filled the pores. Details of the
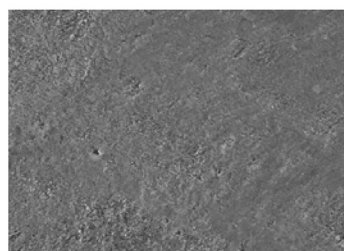

(a) GDL substrate (SGL24BA, d_m=42μm)

(b) Hydrophobic MPL in triple MPL (d_m=3μm)

(c) Hydrophilic MPL in triple MPL (d_m=2μm)

Fig. 2. Surface SEM micrographs of the substrate, hydrophobic MPL and hydrophilic MPL in the triple MPL coated GDL.
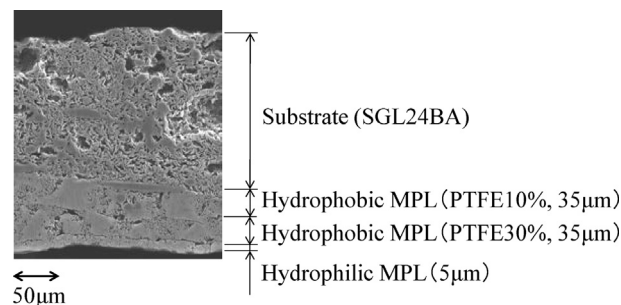

Fig. 3. Cross-sectional SEM micrograph of the triple MPL coated GDL (h_{PTFE} = 70 μm, h = 245 μm).

experimental method may be found in our previous paper [14]. We have previously demonstrated that the best performance under both low and high humidity conditions was obtained with a hydrophobic intermediate MPL pore diameter d_m of 3 μm for the double MPL coated GDL [19]. Therefore, d_m for the hydrophobic MPL in both the double MPL and the triple MPL coated GDLs was set at 3 μm. The hydrophobic MPL was coated on the substrate two times using a bar coating machine. The MPL pore diameter was measured immediately after each coating process. After the first coating, d_m for the hydrophobic MPL was relatively large at 8 μm. The second coating reduced d_m, which became constant at 3 μm for all the hydrophobic MPLs. d_m for the hydrophilic layer in all the double and the triple MPL coated GDLs was set at 2 μm.

Fig. 3 shows a cross-sectional SEM micrograph of the triple MPL coated GDL. The thickness of the GDL substrate (SGL24BA) was 190 μm. When the hydrophobic MPL was coated on the substrate, the MPL was not simply coated on the surface of the substrate, but penetrated into the porous substrate. The boundary between the MPL and the substrate observed in the micrograph was neither clear nor uniform. It was difficult to measure the MPL thickness considering the penetration into the substrate. Therefore, the average thickness of the MPL was evaluated by a comparison of the measured in-plane air permeability using GDLs with and without the MPL [14]. The MPL thickness of the hydrophobic MPL h_{PTFE} was set at 70 μm. The thickness of the hydrophobic intermediate MPL h_{PTFE} in the double MPL coated GDL was also set at 70 μm. For the triple MPL coated GDL, the thickness of the hydrophobic MPL containing either 10 or 40 mass% PTFE after the first MPL coating was set at 35 μm, and that of the hydrophobic MPL containing 30 mass% PTFE content after the second MPL coating was set at 35 μm.
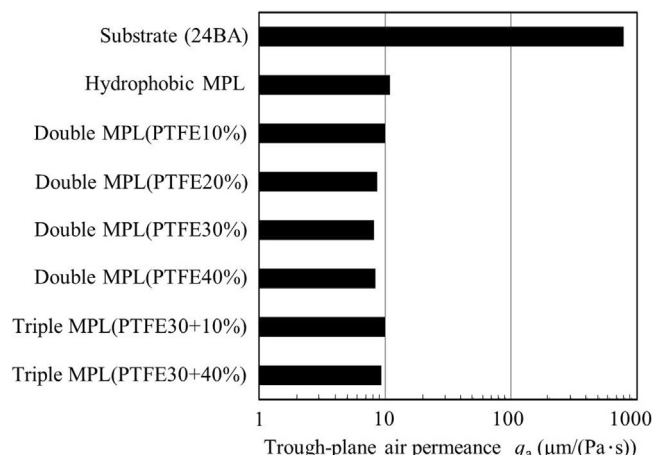

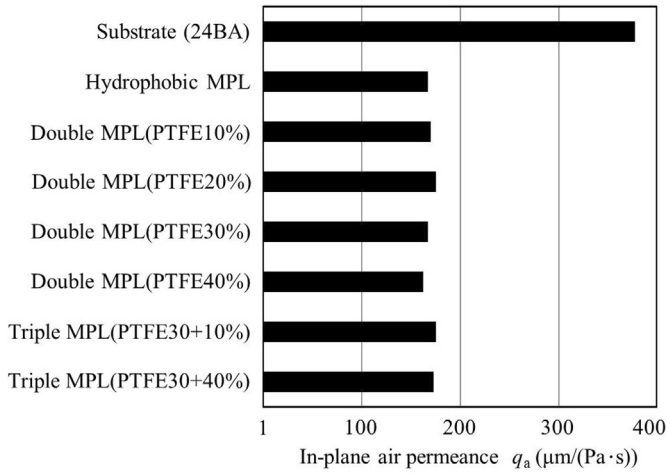**Fig. 4.** Through-plane air permeance of the tested GDLs.

Fig. 5. In-plane air permeance of the tested GDLs.

The total thickness h_{PTFE} of the hydrophobic MPL in the triple MPL coated GDL was set at 70 μm. We have reported that the hydrophilic layer thickness for the double MPL coated GDL should be as small as possible to enhance the PEFC performance without causing flooding in the hydrophilic layer [18]. Therefore, the hydrophilic layer thickness was set at 5 μm and the total thickness h of all the double and the triple MPL coated GDLs was set at 245 μm.

Figs. 4 and 5 show the through-plane and in-plane air permeance q_a obtained for the GDLs with and without MPLs. The through-plane and in-plane air permeances of the GDL were measured using an air permeability test apparatus, as described in our previous paper [14]. The supplied air pressure was set at 1.23 kPa. The air permeance q_a was defined as the flow rate divided by the supplied air pressure and permeable cross-sectional area. The through-plane and in-plane air permeance obtained for all the MPL coated GDLs was significantly lower than that for the 24BA GDL without the MPL. The difference in the air permeance for the hydrophobic MPL, the double MPL and the triple MPL coated GDLs was not significant, because the mean flow pore diameter d_m and MPL thickness h_{PTFE} were set to the same values [14].

Fig. 6 shows the contact angles obtained for the hydrophobic and the hydrophilic MPLs. The contact angle of the GDL was determined under the assumption that the maximum pore diameter of the GDL, measured using the air permeability test, was the same as that measured using the water permeability test [14]. The contact angle for the hydrophilic MPL was 82°, while those for the hydrophobic MPLs were greater than 120°. When the PTFE content
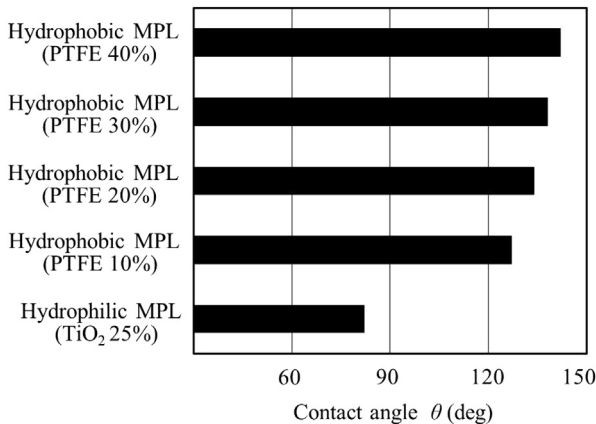
in the hydrophobic MPL was increased from 10 to 40 mass%, the hydrophobicity was enhanced, thereby increasing the contact angle.

3.2. Influence of the PTFE content in the hydrophobic MPL of the double MPL coated GDL on PEFC performance
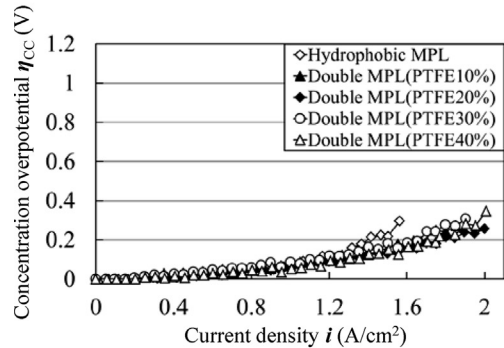
Fig. 7 shows the influence of the double MPL coated GDL used at the cathode on the PEFC performance under low humidity conditions (anode inlet gas: 60% RH, cathode inlet gas: 0% RH) when the PTFE content in the hydrophobic intermediate MPL was varied between 10 and 40 mass%. The parameters for the hydrophobic MPL coated GDL were d_m of 3 μm and h_{PTFE} of 70 μm. The hydrophilic layer for the double MPL coated GDLs was coated on the hydrophobic MPLs, of which the parameters were also d_m of 3 μm and h_{PTFE} of 70 μm. The SGL24BA GDL without the MPL, which was effective at promoting water transport from the humidified anode gas to the MEA, was used at the anode [11]. The output voltages obtained with all the double MPL coated GDLs were higher than that with the hydrophobic MPL coated GDL. The hydrophilic layer in the double MPL was effective at conserving the humidity of the
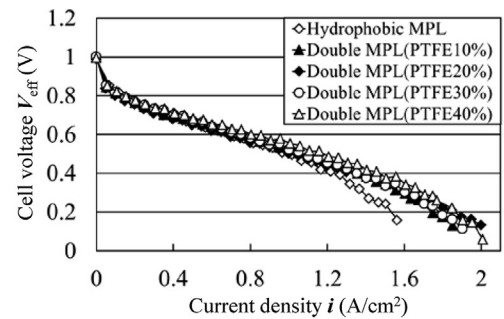
(a) Output voltage
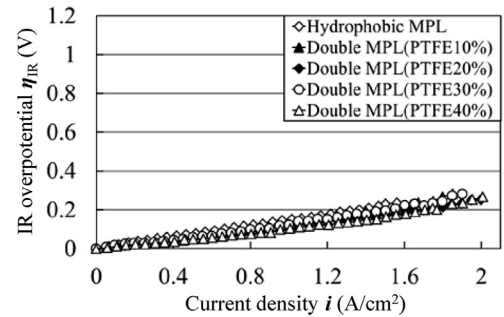
(b) IR overpotential

(c) Concentration overpotential

Fig. 7. Influence of the PTFE content in the hydrophobic MPL ($d_m = 3$ μm, $h_{PTFE} = 70$ μm) of the double MPL coated GDL on PEFC performance under low humidity (Anode: 60% RH, Cathode: 0% RH).
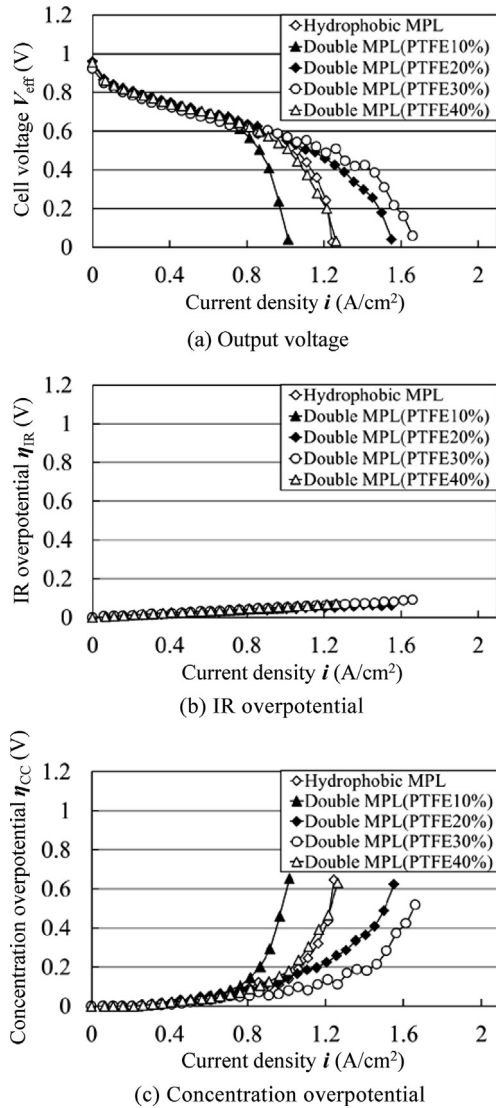

Fig. 6. Contact angles of the hydrophobic and hydrophilic MPLs.

Fig. 8. Influence of the PTFE content in the hydrophobic MPL ($d_m = 3$ μm, $h_{PTFE} = 70$ μm) of the double MPL coated GDL on PEFC performance under high humidity (Anode: 100% RH, Cathode: 100% RH).

MEA, while the hydrophobic intermediate MPL between the hydrophilic layer and the substrate prevented the removal of water from the hydrophilic layer. This resulted in an enhancement of the PEFC performance compared with that for the hydrophobic MPL
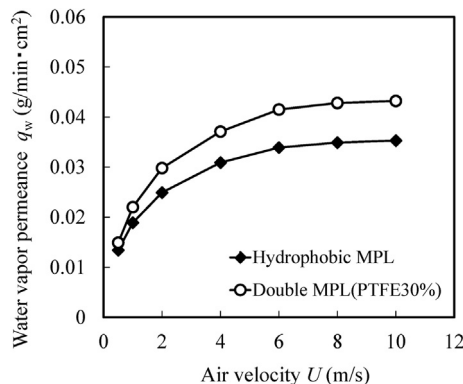

Fig. 9. Variations in the water vapor permeance with increasing air velocity for the hydrophobic MPL and the double MPL (PTFE30%) coated GDLs.

coated GDL [18]. There was no significant difference in the PEFC performance for the double MPL coated GDLs when the PTFE content was varied from 10 to 40 mass%. We have previously demonstrated that a decrease in the PTFE content of a hydrophobic MPL coated GDL helps maintain the humidity of the MEA, which enhances the PEFC performance under low humidity conditions [14]. However, for the double MPL coated GDL, the hydrophilic layer played an important role in conserving the humidity of the MEA. Therefore, the hydrophobicity of the double MPL coated GDL was not a significant factor influencing the ability of the GDL to prevent dehydration of the MEA under conditions where the mean flow pore diameter and thickness of the hydrophobic intermediate MPL were the same.

Fig. 8 shows the influence of the double MPL coated GDL on the PEFC performance under high humidity conditions (anode inlet gas: 100% RH, cathode inlet gas: 100% RH) when the PTFE content in the hydrophobic intermediate MPL was varied between 10 and 40 mass%. The SGL24BA GDL without the MPL was used at the anode. Although the IR overpotentials were almost uniform for all the tested GDLs, the PEFC performance obtained with the double MPL coated GDLs varied significantly with the PTFE content, which is very different to the case for low humidity conditions.

In order to evaluate the effect of the hydrophilic layer in the double MPL, the PEFC performance obtained for the hydrophobic MPL coated GDL was compared with that for the double MPL (PTFE30%) coated GDL. Even though the parameters of the PTFE content, d_m and h_{PTFE} were the same values for both the MPL coated GDLs, the output voltage obtained with the double MPL (PTFE30%) coated GDL was higher than that with the hydrophobic MPL coated GDL. There was no significant difference in the air permeability for both the MPL coated GDLs as shown in Figs. 4 and 5. The water vapor permeability was measured to evaluate the ability of the MPL coated GDL to discharge water from the catalyst layer. The water vapor permeance was measured using the same test apparatus described in our previous paper [19]. Dry air was supplied through the flow channel in contact with the carbon paper substrate. Liquid water was supplied at a pressure of 5 kPa through the flow channel in contact with the MPL. The temperature of the test apparatus was set at 75 °C. The water vapor permeance q_w was defined as the water vapor flow rate divided by the permeable cross-sectional
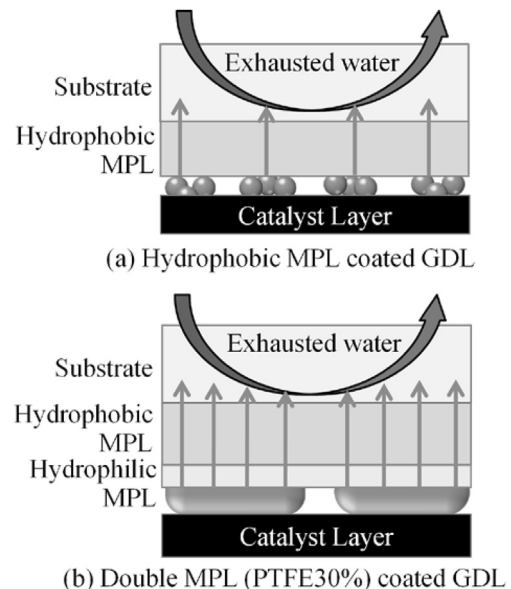

Fig. 10. Comparison of water flow paths from the catalyst layer to the substrate through the MPL for the hydrophobic MPL and the double MPL (PTFE30%) coated GDLs.

area of the GDL. Fig. 9 shows the variation in the water vapor permeance q_w with increasing air velocity U obtained for the hydrophobic MPL and the double MPL (PTFE30%) coated GDLs. The water vapor transport resistance at the GDL/air interface was significantly high under low velocity conditions, which resulted in a low water vapor permeance for both MPL coated GDLs. As the air velocity was increased, the water vapor transport resistance at the GDL/air interface decreased [11], thereby increasing the water vapor permeance. The water vapor permeance obtained for the double MPL (PTFE30%) coated GDL was higher than that for the hydrophobic MPL coated GDL. It was difficult to introduce water into the small hydrophobic pores of the latter GDL, whereas the hydrophilic layer of the former GDL was effective at promoting the introduction of water into the small pores of the hydrophobic MPL. This resulted in a higher water vapor permeability for the double MPL (PTFE30%) coated GDL than for the hydrophobic MPL coated GDL. A double MPL coated GDL with high water vapor permeability could be effective at enhancing the discharge of excess water from the catalyst layer during PEFC operation under high humidity conditions. Fig. 10 shows a schematic diagram that compares the water flow paths from the cathode catalyst layer to the substrate obtained for the hydrophobic MPL and the double MPL (PTFE30%) coated GDLs. It was difficult for water droplets to be spread at the catalyst layer when using the hydrophobic MPL coated GDL, which reduced the water flow paths through the hydrophobic MPL to the substrate. However, the double MPL (PTFE30%) coated GDL promoted water spreading at the catalyst layer, which increased the number of water flow paths for expelling excess water from the catalyst layer to the substrate. This effectively reduced flooding at the catalyst layer, which enhanced the PEFC performance under high humidity conditions.

Fig. 8 also demonstrated that the PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDL had a significant influence on the PEFC performance. When the PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDL was too low, such as 10 mass%, the PEFC performance was lower than that for the hydrophobic MPL coated GDL. Increasing the PTFE content to 30 mass% enhanced the PEFC performance. However, when the PTFE content was as high as 40 mass %, the PEFC performance tended to be degraded. Fig. 11 shows the influence of the PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDLs on the water vapor permeance q_w measured at an air velocity of 10 m s^{-1}. When the PTFE content was increased from 10 to 40 mass%, the water vapor permeance decreased. When the PTFE content was 40 mass%, hydrophobicity of the intermediate MPL was too high, which resulted in a reduction of the water flow paths inside the hydrophobic MPL, thereby
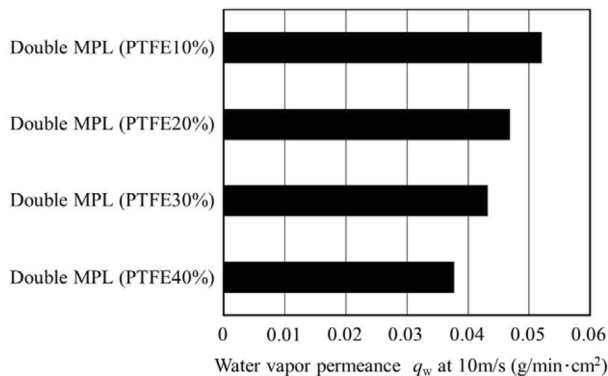
reducing the water vapor permeability. As a result, the discharge of excess water from the catalyst layer to the substrate was inhibited during PEFC operation under high humidity conditions, which increased the accumulation of water at the catalyst layer and promoted flooding, so that no significant enhancement in the PEFC performance could be expected. When the PTFE content was 10 mass%, the introduction of water into the pores of the hydrophobic intermediate MPL was promoted, which resulted in enhanced water vapor permeability of the double MPL coated GDL. However, the hydrophobicity was insufficient for the excess water accumulated in the double MPL to be expelled to the substrate, which caused most of the pores of the hydrophobic intermediate MPL to be filled with liquid water. This inhibited the supply of oxygen from the substrate to the catalyst layer, thereby increasing the concentration overpotential. Therefore, when the PTFE content was too low, such as 10 mass%, no significant enhancement in the PEFC performance could be expected. An appropriate level of hydrophobicity was achieved by increasing the PTFE content to 30 mass %, which was effective at expelling excess water from the catalyst layer through the double MPL to the substrate, while maintaining sufficient oxygen flow paths inside the double MPL from the substrate to the catalyst layer. Therefore, the concentration
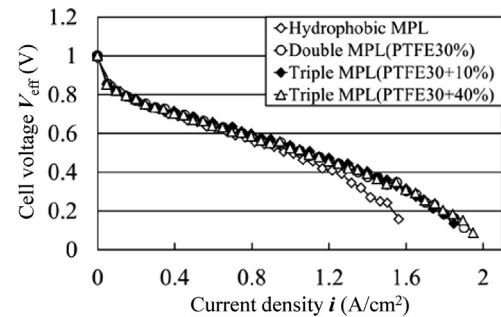
(a) Output voltage
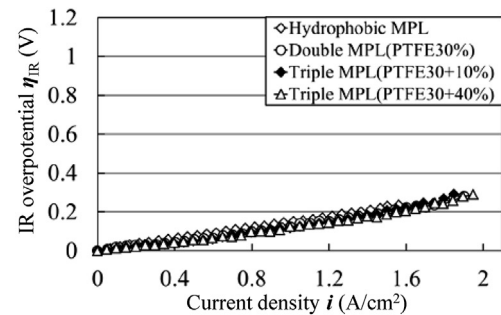
(b) IR overpotential
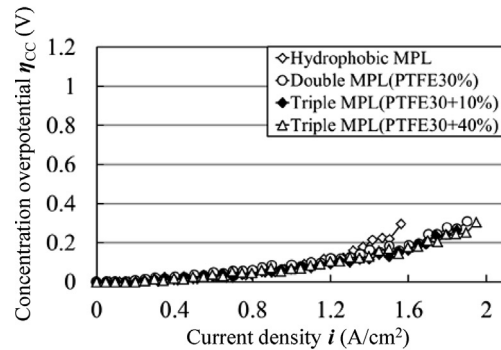
(c) Concentration overpotential

Fig. 12. Influence of the triple MPL (d_m = 3 μm, h_{PTFE} = 70 μm) coated GDL on PEFC performance under low humidity (Anode: 60% RH, Cathode: 0% RH).

Fig. 11. Influence of the PTFE content in the hydrophobic MPL (d_m = 3 μm, h_{PTFE} = 70 μm) of the double MPL coated GDL on the water vapor permeance measured at 10 m s^{-1}.

(a) Output voltage

(b) IR overpotential

(c) Concentration overpotential

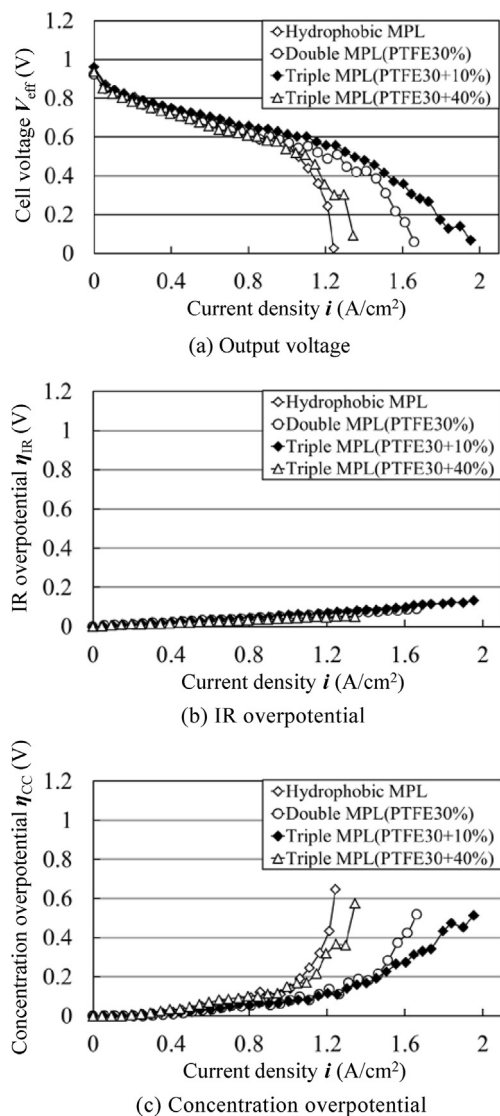**Fig. 13.** Influence of the triple MPL ($d_m = 3$ μm, $h_{PTFE} = 70$ μm) coated GDL on PEFC performance under high humidity (Anode: 100% RH, Cathode: 100% RH).

overpotential was reduced and a higher PEFC performance was achieved under high humidity conditions than that for the hydrophobic MPL coated GDL.

These results demonstrate that the hydrophobicity of the hydrophobic intermediate MPL in the double MPL coated GDL significantly influences the ability to reduce flooding. In the following section, the PEFC performance is evaluated using the triple MPL coated GDL, in which the hydrophobic double MPL has a gradient of hydrophobicity to promote the discharge of excess water from the catalyst layer.

3.3. Influence of the triple MPL coated GDL on PEFC performance

Fig. 12 shows the influence of the cathode GDL on the PEFC performance under low humidity conditions (anode inlet gas: 60% RH, cathode inlet gas: 0% RH). The hydrophobic MPL, the double MPL, and the triple MPL coated GDLs were used at the cathode. The PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDL was set to 30 mass%. For the triple MPL coated GDL, the PTFE content in the hydrophobic MPL in contact with the hydrophilic layer was set to 30 mass% and that in contact with the substrate was set to either 10 or 40 mass%. The parameters for the

hydrophobic MPL in both the double MPL and the triple MPL coated GDLs were d_m of 3 μm and h_{PTFE} of 70 μm. The SGL24BA GDL without the MPL was used at the anode.

The output voltages obtained with the double MPL and the triple MPL coated GDLs were higher than that with the hydrophobic MPL coated GDL. However, there was no significant difference in the PEFC performance between the double MPL and the triple MPL coated GDLs. The hydrophilic layer of both the double MPL and the triple MPL coated GDLs played an important role in conserving the humidity of the MEA. Therefore, the hydrophobicity of the double MPL and the triple MPL coated GDLs was not a significant factor that influenced the ability to prevent dehydration of the MEA.

Fig. 13 shows the influence of the cathode GDL on the PEFC performance under high humidity conditions (anode inlet gas: 100% RH, cathode inlet gas: 100% RH). The hydrophobic MPL, the double MPL and the triple MPL coated GDLs were used at the cathode. The SGL24BA GDL without the MPL was used at the anode. The output voltages obtained for the triple MPL coated GDLs varied significantly depending on the combination of the hydrophobic MPLs. The triple MPL (PTFE30+10%) coated GDL was more effective at enhancing the PEFC performance under high humidity conditions than the double MPL (PTFE30%) coated GDL. However, the PEFC performance obtained with the triple MPL (PTFE30+40%) coated GDL was lower than that with the double MPL (PTFE30%) coated GDL.

Fig. 14 shows the influence of the triple MPL coated GDL on the water vapor permeance q_w measured at an air velocity of 10 m s^{-1}. The water vapor permeability obtained with the triple MPL coated GDLs varied significantly depending on the combination of the hydrophobic MPLs. The triple MPL (PTFE30+40%) coated GDL had lower water vapor permeability than the double MPL (PTFE30%) coated GDL. When the PTFE content in the hydrophobic MPL in contact with the substrate was too high, such as 40 mass%, the water flow paths inside this MPL to the substrate were blocked. This increased the accumulation of water at the catalyst layer and promoted flooding, so that no significant enhancement in the PEFC performance under high humidity conditions could be expected.

In contrast, the triple MPL (PTFE30+10%) coated GDL was effective at enhancing the water vapor permeability. The hydrophobic gradient promoted water transport from the hydrophobic MPL containing 30 mass% PFTE to the hydrophobic MPL containing 10 mass% PTFE, which caused a relatively large amount of water to be stored in the latter MPL in contact with the substrate, which effectively promoted water vapor transport from the hydrophobic MPL to the substrate. Therefore, the triple MPL (PTFE30+10%) coated GDL was effective to expel excess water from the catalyst layer during PEFC operation under high humidity conditions, while
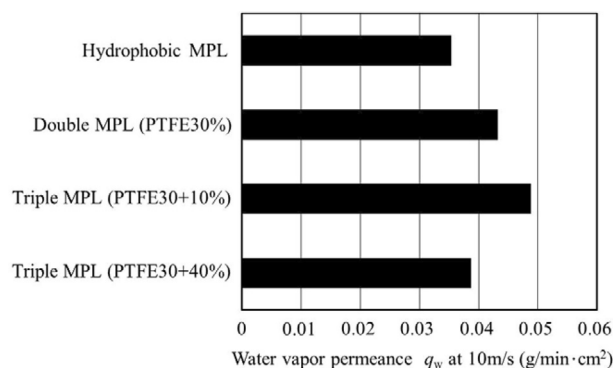

Fig. 14. Influence of the triple MPL ($d_m = 3$ μm, $h_{PTFE} = 70$ μm) coated GDL on the water vapor permeance measured at 10 m s^{-1}.

maintaining oxygen flow paths inside the triple MPL from the substrate to the catalyst layer. This reduced the concentration overpotential and resulted in significantly higher PEFC performance under high humidity conditions than that with the double MPL coated GDL.

In the present study, PEFC performance tests were conducted for 30 h. The polarization curves were stable without any influence of the GDL deterioration phenomena. However, it is also important to evaluate the PEFC performance of the triple MPL coated GDL after long-term durability tests. Further investigations are required to clarify the long-term durability of the triple MPL coated GDL and to evaluate performance enhancements.

4. Conclusions

A novel triple MPL coated GDL, in which a hydrophilic layer was coated on a hydrophobic double MPL, was developed to enhance the PEFC performance under both low and high humidity conditions. The influence of a combination of the hydrophobic MPLs in the triple MPL coated GDL on the PEFC performance was investigated. The following conclusions were obtained:

(1) The hydrophilic layer in the double MPL and the triple MPL coated GDLs was effective at conserving the humidity of the MEA, while the hydrophobic intermediate MPL between the hydrophilic layer and the carbon paper substrate prevented the removal of water from the hydrophilic layer via dry air in the substrate. This enhanced the PEFC performance under low humidity conditions compared with that for the hydrophobic MPL coated GDL. The hydrophilic layer in both the double MPL and the triple MPL coated GDLs played an important role in conserving the humidity of the MEA. Therefore, the PTFE content in the hydrophobic MPL of the double MPL and the triple MPL did not significantly influence the ability of the GDL to prevent dehydration of the MEA.
(2) The hydrophilic layer in the double MPL coated GDL was effective at expelling excess water at the catalyst layer, thereby reducing flooding. This resulted in higher PEFC performance under high humidity conditions than that for the hydrophobic MPL coated GDL. The PEFC performance was more strongly dependent on the PTFE content in the hydrophobic intermediate MPL of the double MPL under high humidity conditions than under low humidity conditions. The best performance was obtained with 30 mass%

PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDL.
(3) The triple MPL coated GDL in which the hydrophobic double MPL had a gradient of hydrophobicity due to variation of the PTFE content, where the PTFE content in the hydrophobic MPL in contact with the hydrophilic layer was set to 30 mass % and that in contact with the substrate was set to 10 mass%, was effective at expelling excess water from the cathode catalyst layer. This resulted in a much higher PEFC performance under high humidity conditions than that for the double MPL coated GDL.

Acknowledgments

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